THE KELLOGG COAL GASIFICATION PROCESS

A. E. Cover, W. C. Schreiner, G. T. Skaperdas

The M. W. Kellogg Company, Houston, Texas   77027

I. INTRODUCTION

The Kellogg Molten Salt Coal Gasification Process represents a unique approach to the problem of coal gasification. The molten sodium carbonate system used for heat supply and catalysis in the Kellogg Coal Gasification Process provides the following advantages. The strong catalytic action for the steam-carbon reaction and coal combustion reaction makes it possible to carry the overall gasification to essentially complete conversion of coal leaving very little fuel to be rejected with the ash. Heat can be supplied by air or oxygen combustion of coal or char from the molten salt, and, in the case of oxygen, it is possible to keep the combustion products separated from the synthesis gas. This has the advantage of keeping sulfur oxides out of the flue gas stream and, in addition, it eliminates carbon oxide dilution of the synthesis gas stream.

Examination of the basic process chemistry and the effect of pressure on the process economics has shown that the Kellogg process is improved by operation at 1200 psia rather than at 400 psia as in the original concept. Operation at high pressure and the separation of the gasification zone from combustion zone makes it possible to take full advantage of the catalytic effect of the molten salt on the direct production of methane from synthesis gas in the gasification step. This is important because it will minimize the heat supply requirement thereby yielding improved economics.

In this process, steam and fine coal are injected continuously into a molten salt bath where they react to form synthesis gas according to Reaction (1):

$$C + H_2O + heat \rightarrow H_2 + CO \tag{1}$$
$$\text{(coal) (steam)} \qquad \text{(synthesis gas).}$$
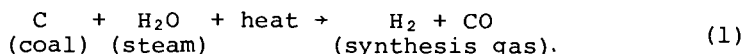
The necessary heat of reaction is supplied by circulating a heated salt stream. In addition, the molten salt mixture is chosen to catalyze Reaction (1) so that it may be carried out at a relatively low temperature. Savings result from the lower reaction temperature, which minimizes the heat demands of the process. The lower reaction temperature also results in a larger concentration of methane in the synthesis gas, which is a decided advantage in the manufacture of pipeline gas. The result is lower investment and lower operating costs.

Since the coal to be gasified is suspended in a salt melt, physical properties of the coal particles are not as critical as in a fixed-bed, moving bed, or fluidized-bed gasifier, where agglomeration and gas channeling must be avoided. It is expected, therefore, that caking, as well as non-caking, coals can be gasified in a continuous manner. Other fuels which can be gasified include petroleum residual fractions (reduced crude), shale, tar sands and municipal waste (garbage)—a brief review of waste gasification has shown this to be feasible. There is, of course, some economic penalty associated with the higher ash contents of some of these fuels. Moreover, better fuel utilization can be obtained using larger particles—hence less grinding equipment and power consumption—than in a suspension gasifier.

The raw synthesis gas produced by the process can subsequently be converted: to a synthetic pipeline gas by catalytic methanation; to hydrogen by water-gas shift and carbon dioxide removal; or to liquid fuels and chemicals via Fischer-Tropsch synthesis.

The molten salt gasification process development program has proceeded in three general areas which include (1) the basic chemistry of the process (2) the testing of materials of contruction to withstand the corrosive, high-temperature environment and (3) engineering feasibility and economic studies.

The present status of the development is illustrated by the following typical results in these three areas. Review of the development work is not complete since time is not available in this symposium to present all the development work.

II. PROCESS STATUS

A. PROCESS CHEMISTRY

The bench-scale process research work was designed to provide the data required to fix the process operation conditions needed to ensure attractive economics. The bulk of the effort was directed toward determining the effects of the major process variables on the rates of gasification and combustion of a broad range of coal feeds. Other work done included viscosity measurements of various melts, melt expansion as a function of gas flow, measurement of salt carry-over, disposition of sulfur in the melt and the development of a recovery procedure for sodium carbonate while removing ash.

The experimental work on gasification and combustion has been carried out in a reactor consisting of a two-inch I.D. × 26 inch long Inconel pipe enclosed in an electric furnace. Variables which have been tested are coal rank, coal particle size, steam partial pressure, temperature, superficial gas velocity, melt depth and ash and carbon content of the melt.

The activation energies for various ranks of coal have been found to vary from 5300 cal/mole for Renner's Cove lignite to 28,600 for anthracite. The gasification rate was found to increase with the 0.55 power of the steam partial pressure and favorable effects on the gasification rate were also found with increasing gas velocity and ash content. The rate is first order with respect to carbon content of the melt but the gasification rate was found to decrease with increasing melt depth. Further gasification tests are planned in the larger, 1200 psi vessel described in the next section on materials testing.

The combustion rate was similarly found to be first order with carbon content but no effect of air pressure or velocity was found. Finer size of coal particles was found to increase the combustion rate and the addition of 0.5% Na_2S increased the rate by a factor of four. Once again, increasing melt depth was found to reduce the combustion rate. The combustion efficiency to CO_2 has been measured to be greater than 85%. Studies on the disposition of sulfur in the process have shown that the flue gas will contain very little sulfur compounds.

B. MATERIALS TESTING

The molten salt system has presented a severe containment problem. Previous work on corrosion of test specimens has shown that Monofrax A (a fused, cast, high-purity alumina) provides satisfactory corrosion resistance and can serve well as a vessel lining. The corrosion tests have included all the components that will be in the reactor under commercial operating conditions, including coal ash, coal sulfur, the sodium-sulfur compounds in the appropriate state of reduction and oxidation as well as coal particles undergoing gasification. In addition corrosion tests of Monofrax A were conducted with test pieces that were spring loaded to compressive stress levels that would exist in the wall of a full-scale commercial vessel. These tests showed clearly that Monofrax A would resist the attack of the reacting system including design stresses in a completely satisfactory manner.

Vessel designs were carried out in sufficient detail to ensure that dependable vessels could be built using a Monofrax A lining. The mortar in the wall will be frozen salt necessitating proper control of temperature gradients through the wall, and designs incorporating such gradient control were found to be entirely practical. It has become necessary to demonstrate the validity of the conclusions from the corrosion test program and the vessel design work and thus to demonstrate the validity of the design for the containment problem. For demonstration purposes, a corrosion test vessel has been designed.

It is expected that this corrosion test unit will, when commissioned, provide proof of feasibility of the commercial design concept. The test vessel chosen is six inches diameter by six feet long and the heat is supplied by combustion of fuel

to produce a simulated synthesis gas. This permits establishment
and demonstration of the temperature gradients essential to the
design, and avoids the very high cost of a melt circulation
system. The fuel chosen is propane which is burned with air at
full process pressure. Steam, hydrogen sulfide and coal ash are
added to the system to simulate the "working" salt conditions
that would be encountered in actual coal gasification.

III. PROCESS DESCRIPTION

Based on the experimental work carried out on the process,
a flow sheet for a plant to produce 262,000,000 standard cubic
feet per day of 956 Btu/SCF pipeline gas has been prepared. The
gasification section of this plant is shown in Figure 1. This
flow sheet design work has had the objective of determining the
economic attractiveness of the process; some items (e.g., coal
feeding) may be modified on further design work. Ground coal
is added to one of the coal lock hoppers and is pressurized with
an inert gas or with compressed synthesis gas. Coal is with-
drawn continously from the pressurized hopper (while the other
hopper is being filled) into to stream of steam at 1500°F and
1250 psia. Steam and coal are injected into the molten salt
gasifier where they are heated by intimate contact with the
molten salt (sodium carbonate) and react according to
Reaction (1) to produce synthesis gas. The synthesis gas leaves
the molten salt bath at about 1750°F and 1200 psia, flows through
a heat exchanger where the sensible heat of the gas is used to
generate steam, is quenched with water and is delivered at 700°F
for further processing.

In the molten salt gasifier where Reaction (1) takes place,
a circulation of molten salt is used to supply the needed heat.
This heat is generated in a separate combustion vessel where
some of the coal in the melt is burned with oxygen which heats
the melt. Melt circulation is accomplished using the gas lift
principle where steam is used to aerate and lift the molten salt
from the combustor to the gasifier. Flow from the gasifier back
to the combustor is by gravity.

The flue gases which heat the melt by direct contact are
generated by combustion of coal with oxygen. Oxygen for this
purpose is compressed to 1230 psia and is preheated against the
flue gases leaving the combustor. The flue gases flow through
the expander which provides the power needed for oxygen compres-
sion and then flow to the ash removal section where the CO_2 is
used to recover the Na_2CO_3.

It should be noted at this point that the two-vessel,
gasifier-combustor system shown in Figure 1 is but one of the
possible designs which are being considered. One alternate is a
single-vessel system where the oxygen is fed directly to the
gasifier. This scheme eliminates the circulating melt but
results in reduced methane production in the gasifier and some-
what higher operating costs.

Ash left in the melt by the combustion and gasification of the coal is allowed to build up to a level of 8 weight percent and settled in a quiescent zone to 16 percent ash. A stream of the ash-carbon-sodium carbonate mixture is continuously withdrawn from the gasifier and flows to ash removal where it is processed to separate the ash from the melt. A simplified flow sheet for this ash removal is presented as Figure 2. The melt stream is quenched to 400°F with a solution saturated with sodium bicarbonate at 100°F in the quench tower. Solid melt particles in the resulting slurry are ground to facilitate dissolution of the salt. This stream is then flashed to essentially atmospheric pressure into a holding tank, where sufficient residence time is provided to dissolve the sodium carbonate. The slurry leaving this vessel is filtered to separate the ash and carbon from the solution. This residue is sent to disposal.

The solution leaving the filter flows to a carbonation tower where the sodium carbonate is reacted with carbon dioxide from the gas purification system. The tower operating temperature is about 100°F. At this temperature the sodium bicarbonate concentration exceeds its solubility limit and it is precipitated from solution. The slurry is then filtered, the bicarbonate leaving the filter is calcined to decompose the bicarbonate to carbonate and is returned to the combustor while the solution is recycled to the quench tower.

Raw synthesis gas delivered from the synthesis section is further processed as shown in Figure 3. The gas is shifted to adjust the ratio of hydrogen to carbon monoxide for optimum methanation, and is then purified and finally converted to methane.

IV. ECONOMICS

The equipment needed to carry out the primary process steps just described as well as all auxiliary equipment needed to supply additional services and controls have been designed for a plant to produce 262,000,000 standard cubic feet of 956 Btu/SCF pipeline gas and the plant cost has been estimated.

Estimated capital investment is summarized in Table 1. Offsite Facilities, Section 1100, encompasses all those facilities needed to make the plant completely self-sufficient, such as fire-fighting equipment, buildings to house administrative and supporting personnel, and equipment to supply steam, electric power and cooling water. Total capital investment for the plant is about $149,000,000. This investment is for a "turn key" plant—that is, one which is completely erected and ready for full operation—and includes the contractor's overhead and profit for the complete engineering and construction of the plant.

Estimated gas manufacturing cost and selling price are shown in Table 2. With bituminous coal charged at a nominal $4 per ton and depreciation at 5 percent per year, estimated gas production cost, before any return on investment, is about 52¢/10^6 Btu. Using the AGA-IGT Utility Accounting Method which assumes 65/35

debt-equity ratio, 5% interest on the outstanding debt, 48% income tax rate and 7% return on the utility rate base, the twenty-year average gas selling price is 62¢/10^6 Btu. With a 7.5% interest rate and 9% return, the gas selling price would be about 65¢/MM Btu.

It is therefore concluded that the Kellogg Coal Gasification Process is an economically attractive process for the production of substitute natural gas. Operation of the gasifier at 1200 psia maximizes the direct production of methane thereby reducing the heat supply requirement (and oxygen requirement) and improving the economics. A comparison has shown that the heat requirement in the gasifier is reduced by 44% at 1200 psia vs. 400 psia. Also, over 99% of the carbon in the coal is converted to synthesis gas owing to the strong catalytic action of the salt on the steam-carbon reaction and the coal combustion reaction, as well as on the coal hydrogenation reaction. For the above reasons, it is felt that further development of this process as an alternate substitute natural gas process or as a synthesis gas or hydrogen producer for hydrogasification or other uses is warranted.
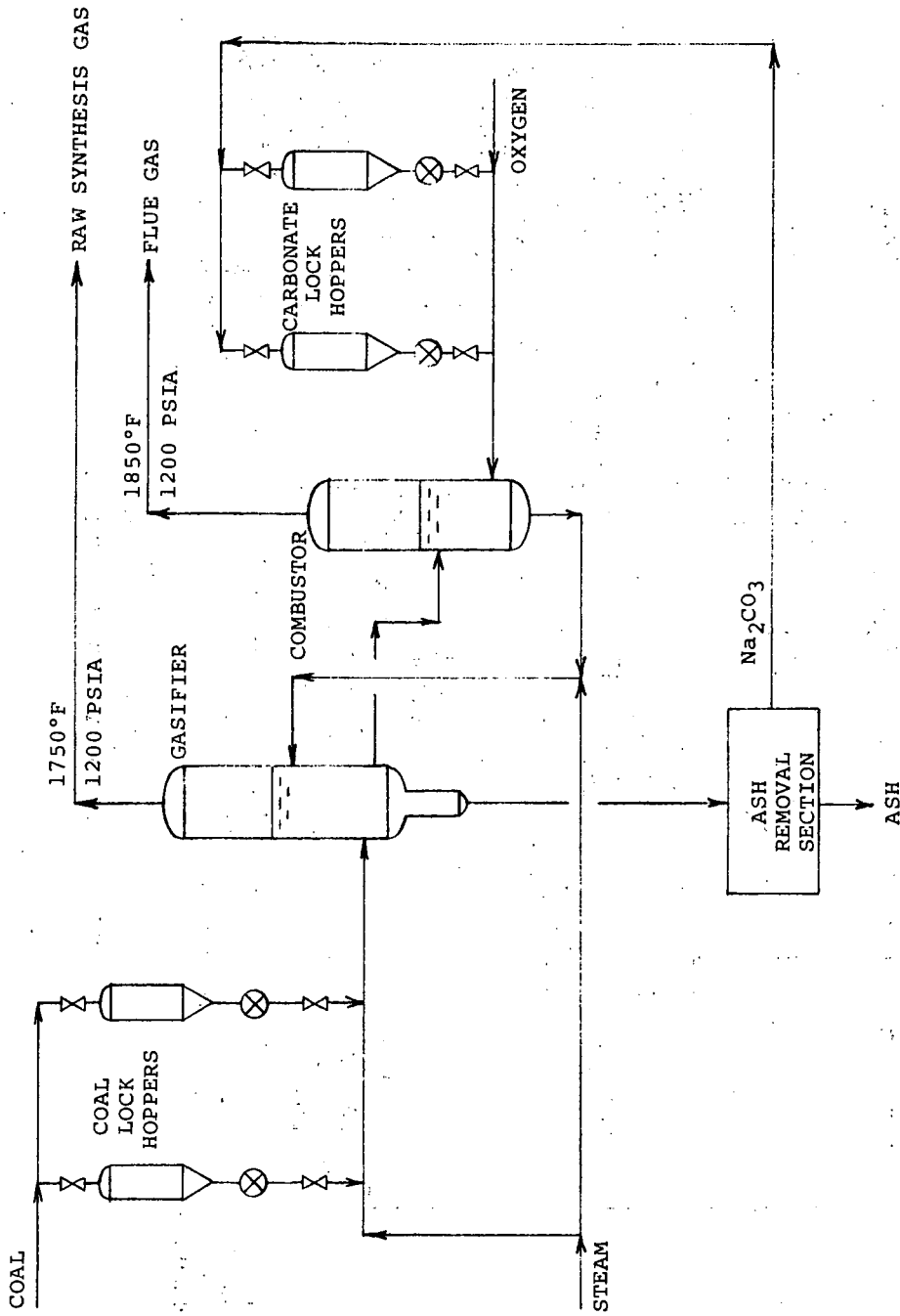
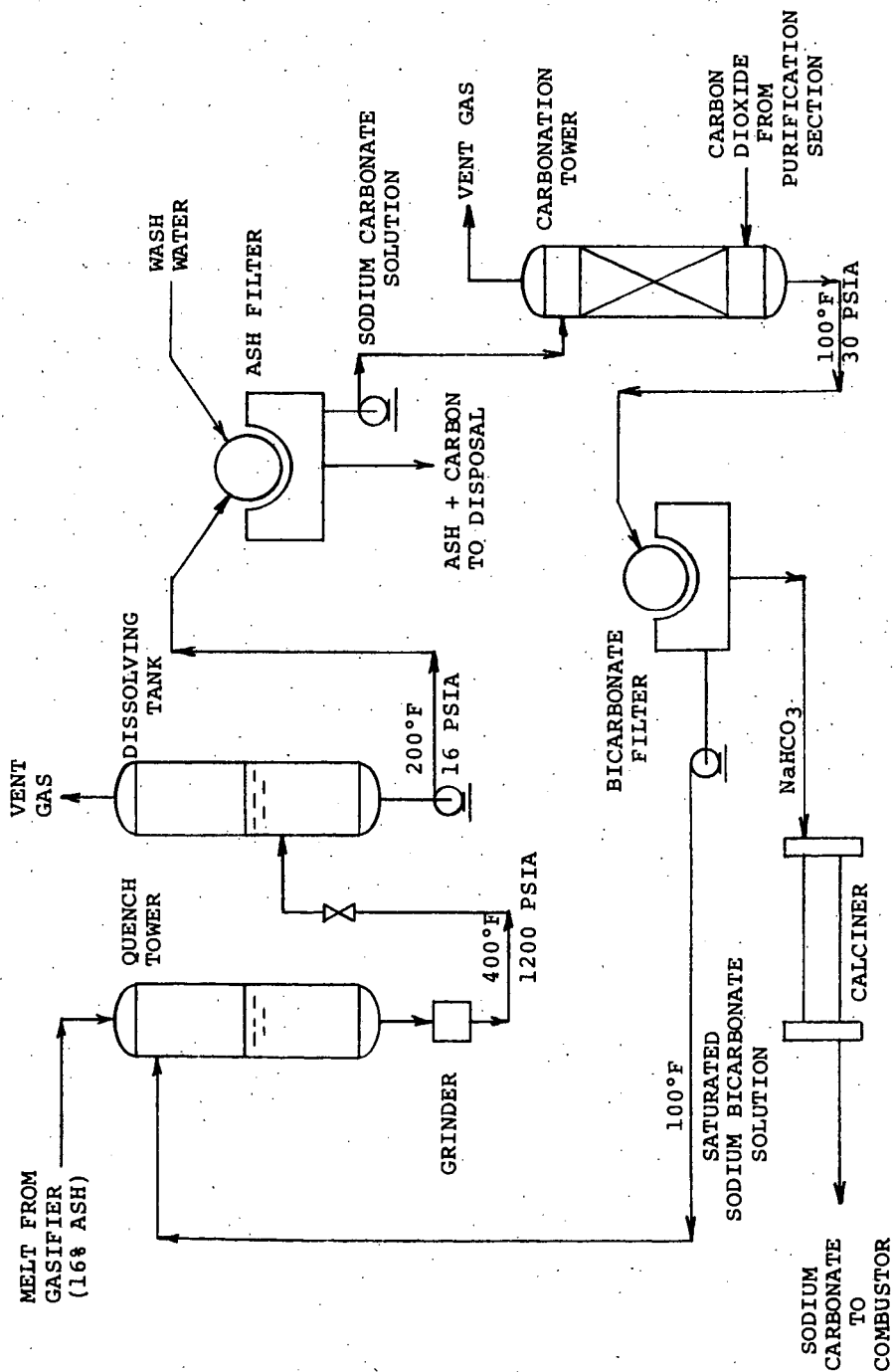FIGURE 1

FLOW DIAGRAM FOR GASIFICATION SECTION

FIGURE 2
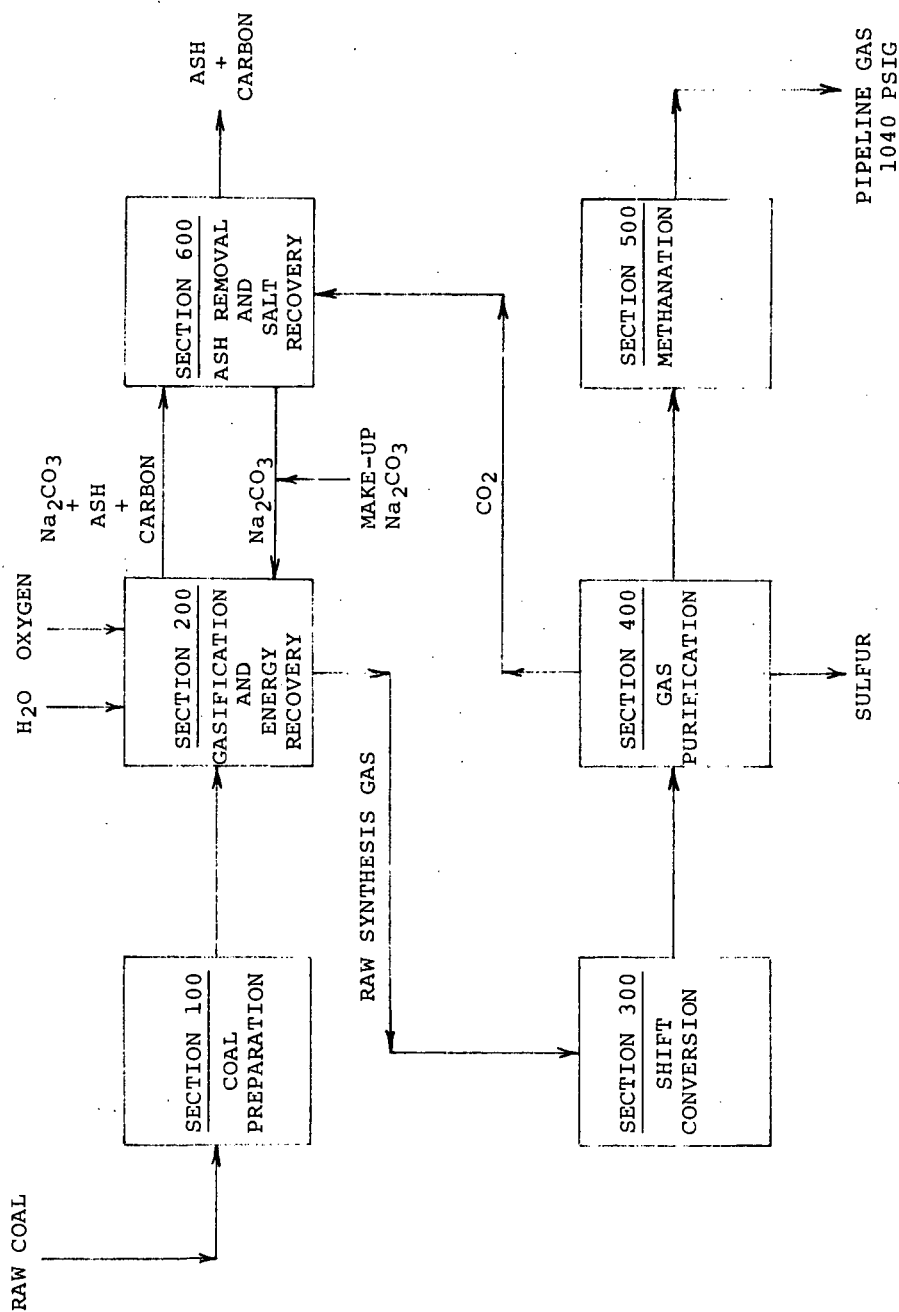
FLOW DIAGRAM FOR ASH REMOVAL SECTION

FIGURE 3

BLOCK FLOW DIAGRAM

PIPELINE GAS FROM BITUMINOUS COAL

TABLE 1

INVESTMENT SUMMARY

PIPELINE GAS FROM BITUMINOUS COAL

KELLOGG MOLTEN SALT PROCESS

Basis: 262,000,000 SCFD of Pipeline Gas
         (250 x 10^9 Btu/day)

Section	Title	INVESTMENT
100	Coal Storage and Preparation	$ 5,100,000
200	Gasification	39,100,000
	Oxygen Plant	30,600,000
300	Shift Conversion	10,000,000
400	Gas Purification	22,100,000
500	Methane Synthesis	4,300,000
600	Ash Removal	13,200,000
1100	Offsite Facilities	18,000,000
	TOTAL FIXED INVESTMENT	$142,400,000
	Working Capital	6,800,000
	TOTAL CAPITAL INVESTMENT	$149,200,000

TABLE 2

ESTIMATED GAS SELLING PRICE

PIPELINE GAS FROM BITUMINOUS COAL

KELLOGG MOLTEN SALT PROCESS

Basis: 262,000,000 SCFD of Pipeline Gas
(250 x 10^9 Btu/day)

Item	Gas Cost ¢/10^6 Btu
Bituminous Coal at $4/ton	17.6
Sodium Carbonate Make-up	2.3
Catalysts and Chemicals	1.0
Sulfur Credit at $10/ton	- 0.4
Power at 6 mills per kwh	6.7
Operating Labor	1.1
Maintenance & Supplies	5.6
Overhead & Supervision	3.8
Depreciation at 5% of Investment	8.6
Local Taxes & Insurance at 3% of Investment	5.2
TOTAL GAS PRODUCTION COST	51.5
GAS SELLING PRICE	61.9